



Article Development of Electrospun Films from Wastewater Treatment Plant Sludge

Gregor Lavrič¹, Aleksandra Miletić², Branka Pilić², Daša Medvešček¹, Saša Nastran³ and Urška Vrabič-Brodnjak^{4,*}

- ¹ Pulp and Paper Institute, Bogišićeva ulica 8, 1000 Ljubljana, Slovenia; gregor.lavric@icp-lj.si (G.L.); dasa.medvescek@icp-lj.si (D.M.)
- ² Faculty of Technology, University of Novi Sad, Bulevar cara Lazara 1, 21000 Novi Sad, Serbia; alexm@uns.ac.rs (A.M.); brapi@uns.ac.rs (B.P.)
- ³ JP Voka Snaga d.o.o., Vodovodna cesta 90, 1000 Ljubljana, Slovenia; sasa.nastran@vokasnaga.si
- ⁴ Department of Textiles, Graphic Arts and Design, Faculty of Natural Sciences and Engineering, University of Ljubljana, Snežniška 5, 1000 Ljubljana, Slovenia
- * Correspondence: urska.vrabic@ntf.uni-lj.si

Abstract: Electrospinning is a versatile method for producing continuous polymer nanofibers, including from wastewater treatment plant sludge (WTPS). In this context, purified WTPS was successfully used to produce electrospun fibers. The main objective of our research was to produce new, local, circular, renewable and environmentally friendly packaging material. The aim of the research was to purify and treat WTPS to make it suitable for the electrospinning process, thus producing a new material and chemically characterizing it in the first step. One of the major advantages of our process was that the electrospinning process could be carried out with water and ethylenediaminetetraacetic acid. The optimal viscosity was determined to be 20,000 mPas in order to produce sufficient nanofibers. Analyses such as Fourier-Transform Infrared Spectroscopy (FTIR) and ¹H-NMR (proton nuclear magnetic resonance) were used to determine the substances of unpurified and purified WTPS. The tensile properties, contact angle, surface properties and differential scanning calorimetry of the final material were determined and used. The ¹H-NMR analysis confirmed the presence of a small quantity of polyhydroxyalkanoates in the samples. Based on the properties, the final material was brittle and less stretchable compared to electrospun packaging films available in the market.

Keywords: wastewater treatment; electrospinning; differential scanning calorimetry; tensile properties; proton nuclear magnetic resonance spectroscopy; packaging

1. Introduction

The extensive consumption of natural resources in recent years is reflected in the increasing consumption of packaging [1], which consequently causes the increase of the amount of plastic waste. According to the World Economic Forum, there will be more plastic than fish in the oceans by 2050 if human habits persist [2]. From a different perspective, the current global health crisis has highlighted the importance of using local and easily accessible raw materials that do not have to travel long distances to reach the consumer. These facts indicate that all future packaging materials should be renewable, environmentally friendly and, if possible, made from alternative raw materials (e.g., industrial waste) [3,4]. In general, waste is described as a product or by-product substance derived from industrial or agricultural processes or other activities with end-use purposes [5]. Waste can be a direct result of processing technology or the product of secondary treatment of waste streams, for example, wastewater, which produces several types of wastewater treatment plant sludge.

Depending on the treatment stage, primary, secondary, tertiary and chemical sludges are produced. Primary sludge is formed during primary treatment (screening, grate removal, flotation, precipitation and sedimentation) when heavy solids, grease and oils are



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). separated from raw wastewater [6,7]. Usually, the primary sludge contains 2–9% solids. The remaining 90% (sometimes 99.5%) is water. Secondary sludge (waste activated sludge) is formed during biological treatment when the biodegradable organic content of wastewater is degraded by microorganisms. The total concentration of solids ranges from 0.8% to 3.3%, depending on the type of biological treatment process, with the remainder being water [7]. The organic fraction of waste activated sludge contains 50–55% carbon, 25–30% oxygen, 10–15% nitrogen, 6–10% hydrogen, 1–3% phosphorus and 0.5–1.5% sulfur [8]. Tertiary sludge is produced in advanced wastewater treatment stages when nutrients (nitrogen and phosphorus) need to be removed [6]. Usually, nutrient removal is carried out simultaneously with organic matter removal. Chemical sludge is produced by chemical processes carried out at the municipal wastewater treatment plant, such as chemically assisted primary sedimentation. In this process, an appropriate coagulant is added to the primary clarifier to reduce the organic load for further biological treatment. The qualitative and quantitative properties of the sludge depend on the reagent used and the dosage. Typical reagents are hydrated lime, ferric chloride, aluminum sulfate and chitosan. Chemical sludge may contain non-negligible quantities of metals, due in part to the inorganic coagulants used. Chemical sludge can also be produced by coagulation-flocculation of the supernatant thickener and by backwashing after sedimentation/chemical-physical treatment [9].

Due to legislation restricting the use of landfills and land as sludge disposal methods, many researchers have attempted to reuse and recycle sludge in the most sustainable possible way [9–25]. Taking into account that the organic components of sludge present a rich source vein in terms of energy and nutrients waiting to be tapped, various studies show that, in the context of the circular economy, an important advantage of energy and fuels derived from waste is that they can replace other energy resources and limit the associated CO_2 emissions. From a scientific point of view, the challenge of sewage sludge is one of the most studied in the last 30 years. The concepts of material and energy recovery, which are milestones of today's circular economy, have already been addressed by several working groups [19–22]. Most of the studies on the material utilization of wastewater sludge are based on the extraction of polyhydroxyalkanoates [9].

Over the years, it has been shown that wastes generated mainly from processes in the agricultural, food, textile and paper industries have a high end-use potential [10]. A good example of high-end potential from the agri-food sector is waste from coffee production, as described by Figueroa et al. and Malara et al. [11,12]. As a waste solution, an anthocyanin-based milk freshness indicator or sensor that could be used as an indicator of actual milk quality was shown by Weston et al. [13]. Researchers have also presented available technologies and materials for exploited cooking oil recycling, which has a significant impact on household waste solutions [14–16]. In recent years, substantial progress has been made in waste processing in the textile and paper industries [17–22].

In the past, studies on the utilization of wastewater treatment plant sludge (WTPS) for various purposes have been published [9,23–25]. Most of them are based on the extraction of polyhydroxyalkanoates (PHAs) from WTPS. PHAs are a class of bio-based and biodegradable polymers produced by bacterial fermentation of complex organic substrates [25]. They belong to the class of polyesters and can be thermoplasts or elastomers, and depending on the structure, they have similar properties to the conventional plastics, which makes PHAs suitable candidates for their substitution [25]. Despite the extensive literature review, no report of the direct use of WTPS for material preparation by electrospinning has been published so far. The materials produced by electrospinning technology are intended for a variety of applications in many fields such as medicine, pharmaceuticals, biotechnology, sustainable engineering materials and even packaging [26–28]. The advantage of the above technology lies in the production of nanofibers and their small diameter, large specific surface area and high porosity. Meanwhile, the rapid development of nanotechnology has enabled new applications for electrospun materials. In general, electrospinning technology is a new strategy for environmentally friendly nanomaterials with special properties and a promising solution for wastewater sludge as well. Accordingly, at least a partial presence of various biopolymers in a local WTPS was expected. With the main objective of producing a new, local, circular, renewable and environmentally friendly packaging material, the sludge was chemically characterized at the beginning of the research. The main advantage of our process is that the electrospinning solution was prepared using water and ethylenediaminetetraacetic acid (EDTA). However, the aim of the research was to clean and treat the wastewater treatment plant sludge in such a way that it is suitable for the electrospinning process, thus creating a new material.

2. Materials and Methods

2.1. Materials

Preparation of wastewater treatment plant sludge solution (WTPS): The WTPS was obtained from the main wastewater treatment plant of Ljubljana. The purification procedure was carried out according to a slightly modified method described by Fang and Jia in 1996 [29]. The purification was based on dissolution of WTPS in a 2% EDTA solution in water and centrifugation. Fifty grams of the absolute dry mass of WTPS was firstly cut down into pieces with a size of ~1 cm × 1 cm. The pieces were then dissolved in a 300 mL EDTA solution and stirred in a 500 mL beaker (500 rpm, 24 h). Centrifugation (16,800 × *g*, 30 min) and separation of the usable supernatant were the final steps of the process. A series of solutions with different viscosities (20,000, 5000, and 2000 mPa) were prepared by water evaporation from the initial solution of WTPS by heating in a water bath (85 °C, ~240 min) while stirring at 250 rpm in order to optimize the process of electrospinning, since it was impossible to obtain continuous nanofibers from the initial solution.

2.2. *Methods*

2.2.1. Electrospinning Process

The electrospinning process was carried out using a Fluidnatek LE -10 electrospinning machine (Bioinicia, Valencia, Spain). Main process parameters: flow rate (μ L/h), needle-to-collector distance (cm) and voltage (kV) were optimized to obtain a satisfactory nanofiber morphology, without visible defects, beads or drops within the fiber structure. The optimal voltage was 19 kV, flow rate was 200 μ L/h and needle-to-collector distance was 15 cm.

2.2.2. Material Characterization

Viscosity Determination

The rheology (viscosity) of the solution was determined using a Viscotech 3000 viscometer (Viscotech Hispania, SL, El Vendrell-España, Spain) according to the Brookfield method. A 20 mL sample was analyzed at room temperature (22 ± 0.5 °C) in an appropriate container providing convenient spindle-to-wall distance.

Fourier-Transform Infrared Spectroscopy (FTIR)

Fourier-transform infrared spectra analysis of WTPS was performed using a Spectrum One ATR-FTIR spectrometer (Perkin Elmer, Waltham, MA, USA). Scans were performed between the infrared regions at $4000-400 \text{ cm}^{-1}$, with an average of 64 scans.

Proton Nuclear Magnetic Resonance Spectroscopy (¹H NMR)

Proton nuclear magnetic resonance spectroscopy of WTPS was performed using an Agilent Technologies DD2 spectrometer (Agilent Technologies, Santa Clara, CA, USA). The WTPS samples were dissolved in D_2O with trifluoroacetic acid (TFA) and deuterated chloroform (CDCl₃) for the purpose of analysis.

Differential Scanning Calorimetry (DSC)

Differential scanning calorimetry of purified WTPS was conducted with a differential scanning calorimeter (TA Instruments Q20, New Castle, DE, USA) within the range of

Tensile Properties

Tensile properties were determined using Zwick Roell Z010 (Zwick Roell, Ulm, Germany) tensile testing machine equipped with a 20 N measuring cell (Class 0.5, ISO 7500-1). The testing speed was set to 1 mm/min. Film strips of 1.5 cm in width and 10 cm in length were used. During the sample stretching, several load and elongation data per second were recorded until the breaking of a sample occurred.

Contact Angle

The wettability and water absorption of material were determined by measuring the contact angle of a 4 μ L water drop using a Fibrodat 1100 (Fibro System AB, Molenbaan, The Netherlands) dynamic contact angle tester. The change in the contact angle in the first 30 s was measured and evaluated.

Surface Characterization with a Scanning Electron Microscope (SEM)

The surfaces/structures of electrospun materials were examined with electron scanning microscopy to acquire information about fiber arrangement and the material network in general. The micrographs were taken with a scanning electron microscope JSM-6060 LV (Jeol Ltd., Akishima, Japan). The instrument was operated at 10 kV at $1500 \times$ magnification.

3. Results and Discussion

In the first part of this study, WTPS obtained from a local wastewater treatment plant was chemically characterized then purified and prepared for the electrospinning process. This process produced a new thin material that could be used for packaging applications. In the final part of the study, the newly obtained material was characterized in terms of tensile strength and wettability.

3.1. FTIR and ¹H NMR Analysis of Wastewater Treatment Plant Sludge

In order to determine the presence of sodium polyacrylate in purified WTPS, FTIR spectra of purified WTPS were analyzed and compared to the sodium polyacrylate spectra (Figure 1).



Figure 1. Fourier-transform infrared (FTIR) transmission spectra (%T) at the wavelength (cm⁻¹) of purified WTPS and sodium polyacrylate.

As shown, three peaks were observed in the pure sodium polyacrylate at 1464, 1571 and 1625 cm⁻¹. The peaks at 1464 and 1625 cm⁻¹ are related to the bending of –CH2– and the C=O stretching mode in the carboxylic acid group of sodium polyacrylate [30,31]. From the spectra, it is evident that purified WTPS and sodium polyacrylate share common peaks at 1464, 1571, and 1625 cm⁻¹ corresponding to the bands of sodium polyacrylate. As previously reported in the literature, the strong peak at 1571 cm⁻¹ remains in the FITR spectra of WTPS, which is also masked by the carboxylate ions of WTPS and sodium polyacrylate [31]. At the same time, in the FTIR spectra of sodium polyacrylate and purified WTPS, the peaks in the range of 1625 and 1800 cm⁻¹ are assigned to the carbonyl groups [30–32]. In the purified WTPS sample, the addition of sodium polyacrylate was detected as expected.

To further analyze the unpurified and purified WTPS, a comparison of the spectra was performed (Figure 2). As can be seen from the spectra, in unpurified WTPS, the peak at 1554 cm⁻¹ is shifted to 1571 in purified WTPS. These two peaks correspond to the carboxylate ions [31]. In purified WTPS, more distinct peaks were observed at 2911, 1362, 1181, and 915 cm⁻¹ wavelengths.



Figure 2. Fourier-transform infrared (FTIR) transmission spectra (%T) at the wavelength (cm⁻¹) of unpurified and purified WTPS.

Figure 3 illustrates the ¹H-NMR of purified WTPS and sodium polyacrylate. Sodium polyacrylate was used for the purposes of WTPS analysis. Therefore, the presence of this component was also detected in the purified WTPS sample. The characteristic solid peaks on the purified WTPS appeared between 1.6 and 2.6 ppm, which are also typical for sodium polyacrylate. A signal at 1.6 ppm belongs to the hydrogen of methylene. The spectra also show the resonance signal CH₂O–COOH bond at 2.580 ppm. As described in the literature on wastewater analysis, peaks between 3.3 and 4.6 ppm could correspond to glycine (3.5 ppm), glycerol (3.6 ppm), serine (3.7 ppm), 2-aminopropanol (3.9 ppm) and 3-hidroxibutyric acid (4.2 ppm) [33]. For the purified WTPS, the strong peaks were detected at 3.7 ppm corresponding to serine and 4.2 ppm corresponding to 3-hidroxibutyric acid.

Figure 4 presents ¹H-NMR spectra of unpurified WTPS and purified WTPS. The strong peak at 1.53 ppm corresponds to water in chloroform. The peaks of unpurified WTPS are stronger, indicating the addition of carboxylic acid in the sample compared to purified WTPS. At the same time, the minor addition of PHA was observed in both samples, as shown in Figure 4. PHA polymers contain hydrogen and carbon; therefore, typical peaks such as CH_2 at 1.35 ppm and CH_3 at 0.85 ppm were detected [34]. Typical peaks for PHA also include CH at 5.2 ppm and CH_2 at 2.55 ppm, which were not detected



in our samples [35]. The peak CH₂ at 1.6 ppm was present but was also masked by the water and chloroform.

Figure 3. ¹H-NMR spectra of purified WTPS and sodium polyacrylate.



Figure 4. ¹H-NMR spectra of unpurified and purified WTPS.

In general, the quantitative estimate of PHA could be determined by the intensity ratio of the signals, and as before, the unpurified sample had higher peaks compared to the unpurified WTPS, especially in the detection of PHA groups. This could be due to the purification process and the reduction of the amount of PHA in the purified sample.

3.2. Determination of Optimal Viscosity and Electrospinning Parameters

The optimal viscosity for the WTPS suspension was experimentally determined to be 20,000 mPas (\pm 500 mPas). This viscosity provided satisfactory nanofiber morphology without visible defects, beads or droplets within the fiber structure (Figure 5 with marked arrows). Purified WTPS with lower viscosities did not lead to uniform structure of the electrospun material. During the electrospinning process of such suspensions, undesirable electrospraying effects occurred in the form of tiny droplets. Higher viscosities (above 20,000 mPas), on the other hand, clogged the electrospinning device system and thus disrupted the production process.



Figure 5. SEM micrographs of final material (**a**) optimal-electrospun from suspension with a viscosity of 20,000 mPas; (**b**) electrospun from suspension with a viscosity of 5000 mPas; (**c**) electrospun from suspension with a viscosity of 2000 mPas at $1500 \times \text{magnification}$.

As described previously, the optimal parameters of the electrospinning process were a voltage of 19 kV, a flow rate of 200 μ L/h and a distance of 15 cm between needle and collector. Only the sample prepared from a suspension with optimal viscosity was further analyzed. Based on the results of the preliminary tests, it shows better performance compared to the other two (sample b and c).

3.3. Differential Scanning Calorimetry Characterization of the Final Material

The DSC heating curve is shown in Figure 6. The analysis was done in one heating cycle only, since the nanofiber morphology of the material is disrupted by the heating and further cooling, reheating serves no purpose. A small peak on the derivative curve at 62.60 °C indicates the glass transition temperature of the new material. PHAs usually have a bit lower glass transition temperature, but since this material contains a considerable number of impurities, the increase might be a consequence. Low-intensity peaks around 130 and 137 °C might also indicate the presence of impurities in the final material. The melting point of the material was detected at around 150 °C, which is in accordance with the research published by Lorini et al. [36].



Figure 6. DSC of purified WTPS.

3.4. Tensile Properties and Contact Angle Analysis of Final Material

The results of the tensile analysis and contact angle analysis of the final material are summarized in Table 1 and presented in Figure 7. As can be seen from the results, the sample exhibited lower stress and strain than the results obtained in the literature [24–26]. The results confirmed a brittle sample, as only 0.422 N/mm² was applied to break it. The same trend was observed for strain, which was only 2.07%. This was more than 50% lower compared to the literature results. The tensile tests confirmed that purified WTPS was suitable for performing electrospun material, but it was very brittle. Therefore, one of the solutions could be a combination with other waste biopolymers or recycled polymers to obtain flexible material in further research.

Table 1. Stress-strain with standard deviation and contact angle measurements for final material.

Analysis Parameters	Sample	Results from the Literature: Bio-Based Electrospun Materials [37–39]
Stress (N/mm ²)	0.422 ± 0.053	1.5–1.85
Strain (%)	2.07 ± 0.129	4.71-13.8
Contact angle	0°	0.4° – 22°

As shown in Table 1, the newly produced material reached a meagre value of the contact angle determined with water at a time of 1 s (0°). According to the solubility of the base material (WTPS) in the presence of EDTA in water, such a material property was expected. The presence of many accessible OH groups on the surface, in combination with other properties (size and interweaving of electrospun fibers), resulted in a material with high wettability. This property could prevent it from being used for packaging applications. However, with an appropriate blending method and/or surface treatment or coating, this could be drastically improved or adjusted. Since we wanted to present the properties of material derived only from WTPS in this study, these options were not used. Other chemicals that could improve this and possibly other properties of the material were also not used due to the preference for developing the new material in the most sustainable and environmentally friendly way possible. In the further development of the material,



methods such as plasma and UV treatment and the possible use of other substances will also be used in accordance with these principles.

Figure 7. Stress–strain curve of the final material.

4. Conclusions

Biopolymers are currently in the spotlight in terms of their production and use, which is continuously increasing. This work evidences that it is possible to produce electrospun fibers from purified WTPS by using water and EDTA. Firstly, the viscosity was determined at 20,000 mPas, which led to satisfying nanofiber morphology, without visible defects, beads or drops within the fiber structure. Due to the presence of impurities in the WTPS, the glass transition temperature of the material was found to be at a higher temperature than usual for PHA, along with two low intensity peaks below the melting temperature. The melting point at 150 °C is within the range of temperatures characteristic for PHA polymers. The low amount of PHA was also confirmed in both samples. Due to the process of purification, the amount of PHA was lower in purified WTPS. However, all the analyzed properties showed that the final material was brittle and less stretchable compared to the packaging films on the market. Further research should be conducted on the possible addition of other biopolymers produced from alternatively sourced or recycled materials to purified WTPS to obtain a material with desired mechanical properties.

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